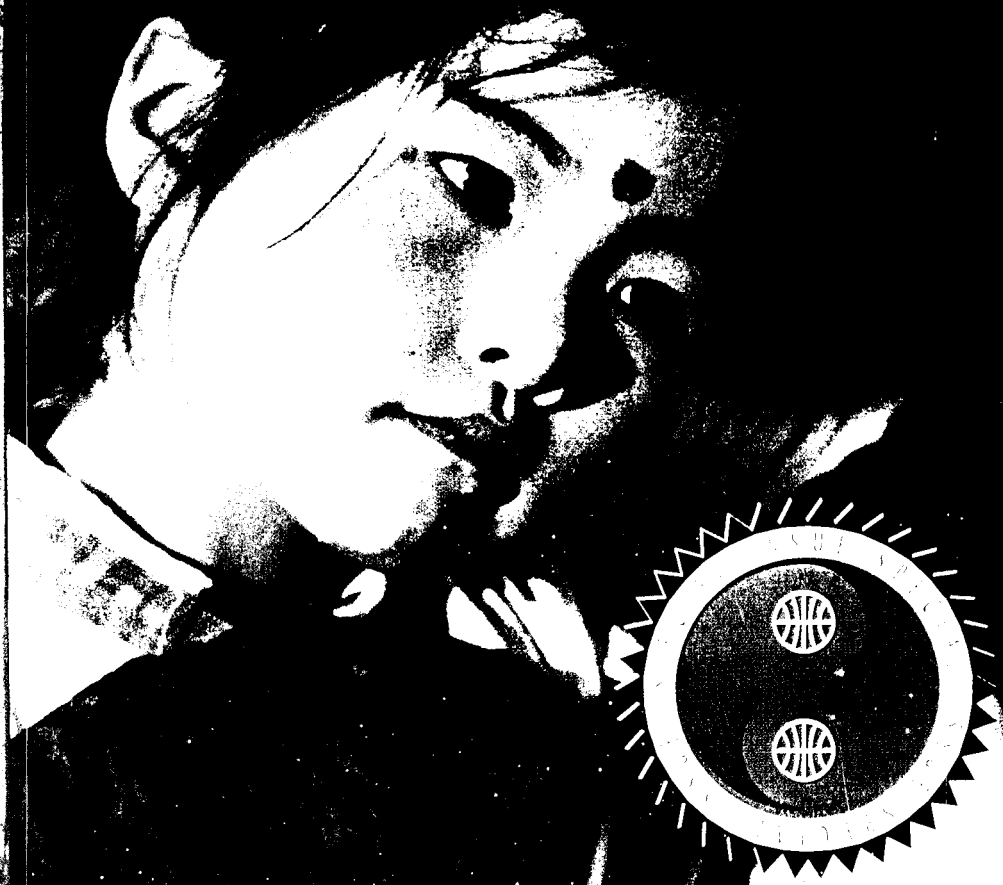


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Evidence for Radionuclide Transport and Mobilization in a Shallow, Sandy Aquifer

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Evidence is reported for the movement of plutonium, americium, thorium, and radium in a shallow, sandy aquifer after the forced injection of colloidal and macromolecular natural organic materials (humic and fulvic acids). Ultrafiltration was used to size-fractionate the materials smaller than 0.45 μm in the injection water. Characterization of these organic materials showed the most mobile to be primarily fulvic acids with a high carboxylate content. Fallout-derived plutonium and americium in the injected materials were transported in the aquifer with the smaller organic fractions. Americium was shown to move from the less mobile colloidal materials to the smaller more mobile fulvics during transport. Thorium, uranium, and radium levels all increased upon injection of the organics into the aquifer, demonstrating the ability of low molecular weight, high carboxylic content fulvic acids to dissolve and mobilize radionuclides from the aquifer's mineral matrices. This effect also increased with decreasing flow rate in the aquifer. The implications of these observations for the interactions of low-level radioactive waste with natural organics are discussed.

Introduction

It has become increasingly apparent that natural humic and fulvic acids present in groundwaters can act as strong complexing agents for metals and radionuclides (1-6). These natural organic species range from macromolecular to colloidal in size and, therefore, are able to move with the groundwater, carrying with them the complexed metals (7-9). Most groundwater models of radionuclide adsorption and transport only include simple complexing agents and the mineral matrix when attempting to predict the movement of actinides. The recognition that colloidal material and macromolecular species smaller than 0.45 μm can act as mobile complexing agents and absorptive particles has led to the proposal of a three-phase model for radionuclide transport (10-12). Humic and fulvic acids compose an important fraction of the active organic complexing agents in most groundwater systems (1-12).

Evidence suggests that carboxylate functional groups are the most active complexing sites for metal binding by humic and fulvic acids. These same carboxylate functional groups give the macromolecular materials their aqueous solubility. Indeed, the ability of humic and fulvic acids to promote mineral dissolution is similar to that observed for the simpler organic carboxylic acids (13-16). Humic materials that are high in carboxylate content, therefore, may also mobilize radionuclides which are trapped in the subsurface mineral matrices.

Plutonium and americium have been reported to be transported over appreciable distances (>3 km) in a semiarid aquifer in volcanic tuff (17), presumably because of complexation by macromolecular or colloidal material.

In this work, we have examined the potential for actinide transport during forced injection of natural organic material into a shallow, sandy aquifer. Ultrafiltration techniques were used to characterize the colloidal and macromolecular materials in the injection water and the baseline aquifer waters with regard to their humic and fulvic acids, inorganic composition, and radionuclide content. Radionuclides determined included the fallout-derived plutonium and americium and the naturally-occurring uranium, thorium, and radium.

Data presented here demonstrate that plutonium and americium, associated with small molecular weight organic materials in surface waters, will migrate in the subsurface environment, consistent with previous observations (17). In addition, naturally-occurring uranium, thorium, and radium appear to be dissolved by these organics from the sandy substrates in the aquifer and are also mobilized. The implications of these results with regard to low-level waste storage and containment are discussed.

Experimental Section

Background A two-well injection experiment was performed in the summer of 1990 in an unconfined, sandy aquifer in a coastal plain located in the Baruch Forest Science Institute, near Georgetown, SC. The site and the experiment have been described in detail elsewhere (18-21). Briefly, surface water with a high dissolved organic carbon (DOC) content (>60 ppm DOC) was obtained from a nearby wetlands area and transferred to an enclosed Viton-lined holding pond near the injection site. After particulate matter (>0.45 μm) was removed, this organic-rich water was injected into the unconfined sandy aquifer and withdrawn from a second well 5 m away. A forced gradient was established prior to injection by recirculating water from the withdrawal well to the injection well at a flow rate of 3.7 L/min. During DOC injection, water from the withdrawal well was discarded downgradient from the injection well. For the purpose of this study, samples were obtained for the determination of radionuclides after initial breakthrough of the DOC at a sampling well located 1.5 m downfield from the injection well. Three sampling ports were located at depths of 1.2, 1.8, and 2.7 m below the surface and designated as A1, A2, and A3, respectively.

The aquifer itself is about 3 m thick and is bounded at this depth by an impervious clay layer. It exhibits three distinct horizons within the study site. Below the unsaturated soil is a 1-m layer of fine brown sand (A1), below this was a region of fine gray sand (A2), and finally a layer of coarser brownish sand (A3). The sampling depths used in this study corresponded approximately to the midpoints of each layer. Each of the three zones exhibited different flow rates under the forced gradient. These were estimated at 477, 178, and 636 cm^3/min for A1, A2, and A3, respectively (18). The redox potential of the groundwaters prior to injection was 142 mV (relative to the standard hydrogen electrode). Dissolved oxygen was <0.1 mg/L (18).

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Sample Filtration. Background samples were obtained from the aquifer and injection water prior to the injection experiment. Subsurface samples were also obtained at three depths (1.2, 1.8, and 2.7 m) immediately after the initial breakthrough of the organics in each of the three layers, as indicated by a sudden rise in dissolved organic carbon content. After the samples were prefiltered with 0.45- μ m membrane filters, hollow fiber ultrafiltration was performed using Amicon filter cartridges of 0.1 μ m and 100 000, 30 000, 10 000, and 3000 molecular weight (MW) pore sizes. The small fraction was then filtered with a 500 MW flat-disk membrane filter (Spectrum Medical Industries) in a vortex mixing stirred cell (Amicon Model 402).

Humic and fulvic acid contents in the size-fractionated samples were determined by adjusting the pH of the samples to 2.5 and filtering the precipitated humic acids. Gravimetric analysis was used to determine the relative contributions of the humic and fulvic acids and the amounts of organic (volatile) and inorganic (nonvolatile) materials present.

Analytical Methods. Sample preparation procedures for radionuclide analysis have been reported elsewhere (22, 23). Low-background α counting was performed to determine ^{232}Th , ^{238}U , ^{226}Ra , ^{228}Ra , ^{241}Am , and $^{239,240}\text{Pu}$ using a multichannel pulse height analyzer (Ortec Model 918A) coupled to a microcomputer (IBM PC). The surface barrier detectors (Paul Downey and Co., Model No. P450-21-100B) used for α measurements have a resolution of <21 keV. Recorded backgrounds were 0.2 count/1000 min. The system configuration yields an efficiency of 34% for measurements of α particles. Results are reported in milli-Bequerels (mBq).

Concentrations of trace metals were determined by inductively coupled plasma (ICP) spectroscopy (Instruments SA Model JY 86, with an HF-compatible torch). Organic carbon contents were measured using a DOC analyzer (Sybron photochem organic carbon analyzer, Model E3500) and are reported as ppm DOC. Humic materials were characterized by pyrolysis gas chromatography (Shimadzu GC-Mini-2 with a PYR-2A pyrolyzer attachment) and Fourier transform infrared spectroscopy (Mattson Polaris with a cooled Hg/Cd/Te detector). Infrared measurements obtained on dried samples by diffuse reflectance (Spectra Tech DRIFT accessory) techniques are reported in Kubelka-Munk units. Infrared absorption measurements were also obtained directly on aqueous samples by using cylindrical internal reflectance (Spectra Tech CIRCLE accessory). Detailed experimental procedures appear elsewhere (24, 25).

Results and Discussion

Background Sample Characterization. Samples of groundwater were obtained from each subsurface layer before the injection of organics and analyzed for trace metals and radionuclides. These results are shown in Table I. All background groundwaters contained a low concentration (approximately 0.5 ppm DOC) of very small organics, most likely fulvic acids (8). Filtration analysis of these subsurface organics determined that all had MW less than 3000 with 40% (0.2 ppm) less than 500. Subsurface sands obtained from core samples near the injection well were similarly analyzed for metals and radionuclides. The results are given in Table II. The

Table I Total Concentrations of Dissolved Organic Carbon, Radionuclides, and Trace Metals in Injection Water (I) and Subsurface Water at Depths of 1.2 (A1), 1.8 (A2), and 2.7 m (A3) before Injection

	I	A1	A2	A3
DOC, ppm	58.0	0.45	0.55	0.50
pH	6.8	6.9	6.7	7.1
plutonium-239, mBq/L	0.137	0.0	0.0	0.0
americium-241, mBq/L	0.058	0.0	0.0	0.0
thorium-232, mBq/L	0.77	0.13	0.40	0.52
uranium-238, mBq/L	1.43	0.140	0.341	0.262
radium-226, mBq/L	0.35	1.04	1.67	0.36
radium-228, mBq/L	0.13	2.22	2.20	2.24
calcium, ppm	19.2	15.5	22.6	25.7
magnesium, ppm	1.4	0.7	1.0	1.1
strontium, ppb	60	69	93	111
aluminum, ppb	55	13	41	<10
iron, ppm	0.7	5.6	5.1	1.4
silica, ppm	2.1	5.0	4.5	4.6

Table II Radionuclide and Metal Compositions of Subsurface Sands at Depths of 1.2 (A1), 1.8 (A2), and 2.7 m (A3)

	A1	A2	A3
thorium-232, mBq/g	2.52	6.62	1.41
uranium-238, mBq/g	1.00	2.89	0.81
radium-226, mBq/g	2.04	5.59	1.31
calcium, ppm	95.7	235.9	70.6
magnesium, ppm	85.5	250.1	39.3
strontium, ppm	2.22	6.45	1.79
aluminum, ppm	2344	7412	965
iron, ppm	3425	2668	3887
titanium, ppm	195.4	195.9	64.3
manganese, ppm	7.5	14.2	11.2

Table III Dissolved Organic Carbon, Trace Metal, and Radionuclide Size Distributions in Injection Water*

	fraction			
	1	2	3	4
DOC	1	13	52	34
plutonium-239	42	0	53	5
americium-241	44	0	50	6
thorium-232	28	6	60	6
uranium-238	39	8	45	8
radium-226	0	0	25	75
calcium	9	0	30	61
magnesium	6	0	24	70
strontium	0	0	33	67
aluminum	25	11	54	10
iron	22	16	58	4
silica	2	0	2	96

* Size fractions are (1) 0.1–100 000, (2) 100 000–30 000, (3) 30 000–3000, and (4) <3000 molecular weight. Expressed as percent of total concentration.

major components of all three sand samples were silica, aluminum, and iron. Concentrations of silica, the bulk constituent, are not shown in Table II.

Table I also gives the total concentrations of trace metals and radionuclides for injection water after filtration with a 0.1- μ m cartridge filter. Table III shows the molecular size distribution of each species in the injection water, expressed as percent of the total concentration. Most of the organics in this surface water had MW values less than 100 000 with 86% less than 30 000. These smaller fractions are also the most active, binding 76% of all species measured. A portion of the radionuclide (38%), along with some aluminum and iron (23%) also appear in the larger size fraction (>100 000 MW), although little organic

Table IV. Characterization of Organic Materials in Mobile Size Fractions of Injection Water and Subsurface Waters at Depths of 1.2 (A1) and 1.8 m (A2) after Injection*

	injection			breakthrough	
	2	3	4	A1	A2
% humic acids	19	2	2	4	3
% fulvic acids	81	95	98	96	97
carboxylate content (mequiv/g)	14	5	5	11	10
% bound carboxylate	71	56	50	63	67
% free carboxylate	29	44	50	37	33
% volatiles					
humic fraction	88	78	72	75	83
fulvic fraction	59	71	73	75	80

* Size fractions are (2) 100 000–30 000, (3) 30 000–10 000, (4) 10 000–1000 molecular weight.

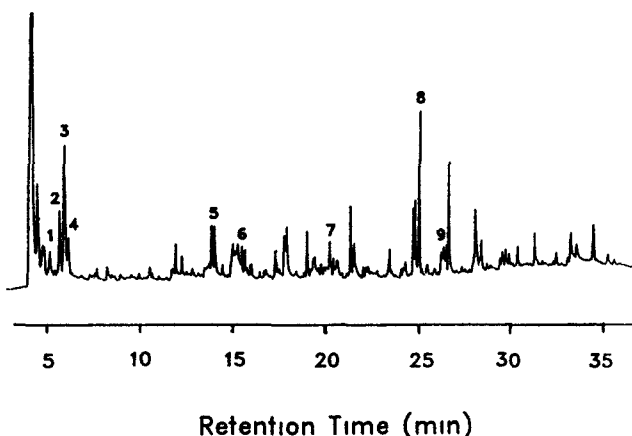


Figure 1 Pyrolysis gas chromatogram (pyrogram) of organic materials isolated from surface water used for injection into the sandy aquifer. Major signature compounds are (1) benzene, (2) acetonitrile, (3) thiophene, (4) toluene, (5) acetic acid, (6) pyrrole, (7) acetamide (8) phenol and *o*-cresol, (9) *p*-cresol and *m*-cresol

material was present. This fraction was therefore primarily composed of larger inorganic colloidal materials

Organic Characterization. The results of gravimetric analyses (Table IV) show that the organic materials isolated from the injection water were 81–98% fulvic acids (i.e., soluble at all pH values) which are composed of 29–41% nonvolatile components

Pyrolysis gas chromatography (GC) was used to determine the distribution of organic functional groups among the size fractions. A typical pyrogram is shown in Figure 1. Unlike other surface waters studied by this technique (24), all size fractions below 100 000 MW resulted in the same signature pattern, indicating that all the organics had the same functional group distributions regardless of molecular size. The organics are therefore expected to exhibit similar chemical properties throughout the size ranges. In general, the pyrogram is also much less complex than those reported earlier (24), suggesting that these natural organics possess more regular and simpler structures than other surface waters studied by this method. The major signature compounds present in the pyrogram are phenol and cresols (indicating a lignin-type backbone structure), benzene, toluene, and acetic acid (indicating a relatively large percentage of both aromatic and aliphatic carboxylates). The sensitivity of this technique is poor, however, for the quantitative determination of aliphatic carboxylate because substantial decarboxylation can occur during pyrolysis. The lack of complexity in the pyrogram

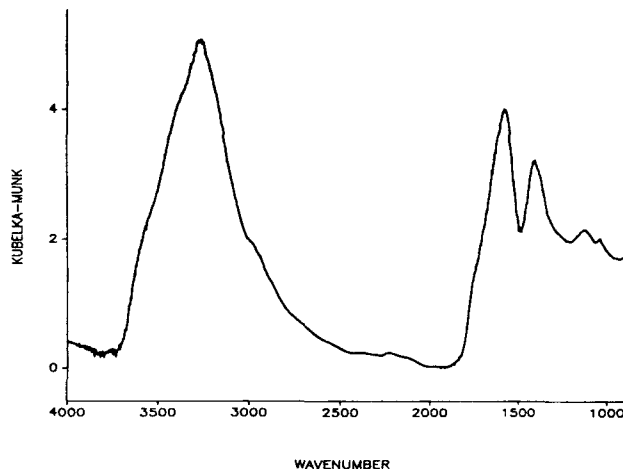


Figure 2 FTIR diffuse reflectance spectrum of organic materials obtained from surface water used for injection into the sandy aquifer

suggests that the carboxylate may be the major functional group present in these organics

A method better suited for the study of carboxylate content of humic materials is Fourier transform infrared (FTIR) spectroscopy. A diffuse reflectance spectrum of the organics isolated from the injection water is presented in Figure 2. Again, the only prominent features present are those assigned to the carboxylate stretch (1585 and 1410 cm^{-1}) with a small contribution from polysaccharide units at 1130–1042 cm^{-1} . As with pyrolysis GC, results obtained from diffuse reflectance analysis were similar for all size fractions

Figure 3 presents FTIR spectra of the size-fractionated injection humics in aqueous solution, obtained with cylindrical internal reflectance techniques. Examination of the humic materials in their natural dissolved state reveals some differences between the size fractions. The hydrated carboxylate bands now appear at 1575 and 1395 cm^{-1} , a decrease of 10 and 15 cm^{-1} from their location in the solid state spectrum. An additional feature which appears at 1620–1630 cm^{-1} and is not present in Figure 2 is a result of a shifting of the carboxylate stretch (1575 cm^{-1}) due to strong association with metal cations in the aqueous phase (26)

It has been demonstrated experimentally that the number of carboxylate groups in a humic or fulvic acid can be quantitatively determined from the integrated band strengths of the carboxylate stretch at 1575 cm^{-1} (25–28). In addition, the ratio of the band intensity at 1620 cm^{-1} to that at 1575 cm^{-1} will give a measure of the number of carboxylates which are directly coordinated to metals in solution. Results of this analysis are given in Table IV.

Characterization of Breakthrough Water. Samples were obtained from the aquifer after the initial breakthrough of the injected humic materials. The DOC concentrations in each size fraction from these samples are shown in Figure 4. The largest colloidal fraction (>100 000 MW) was not present in any of the three breakthrough samples. However, as reported previously (18), the smaller size fractions showed no dramatic change in size distribution patterns in comparison to the original injection water (less than 100 000 MW), except for a small (3–5%) decrease in the 100 000–30 000 MW size range. This observation is in agreement with the structural similarities among the organics from each size range. Most

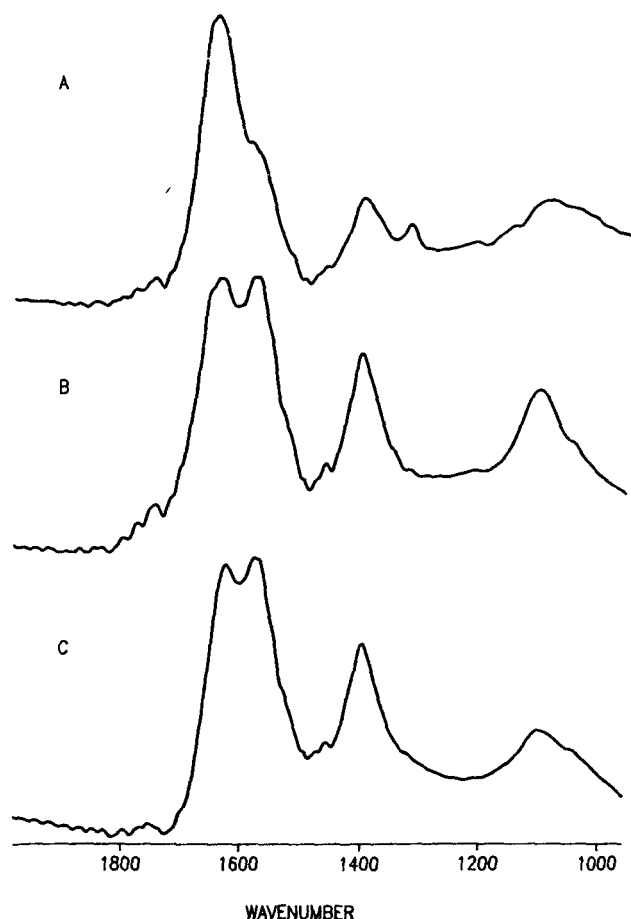


Figure 3. FTIR cylindrical internal reflectance spectra of aqueous solutions of size-fractionated organic materials obtained from injection water. Size fractions are (A) 100 000–30 000, (B) 30 000–10 000, and (C) 10 000–1000 MW.

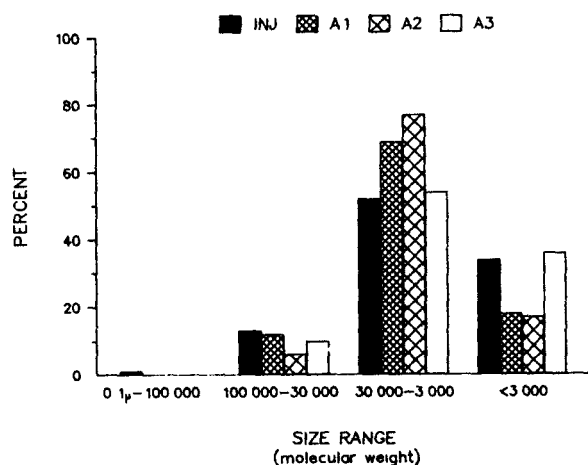


Figure 4. Dissolved organic carbon (DOC) concentrations in size fractions of injection water (INJ) compared to samples obtained from the aquifer at depths of 1.2 m (A1), 1.8 m (A2), and 2.7 m (A3) after injection.

of the humics moving through the aquifer at all levels were less than 30 000 MW.

Table IV also shows that the larger, less mobile organics (100 000–30 000 MW), although high in carboxylate content, had a larger percentage of bound carboxylates prior to injection and were higher in humic acid content than the smaller fractions. In contrast, the organics obtained in the initial breakthrough samples were higher in average carboxylate content than the same size fractions before

Table V. Total Concentrations of Dissolved Organic Carbon, Radionuclides, and Trace Metals in Subsurface Water at Depths of 1.2 (A1), 1.8 (A2), and 2.7 m (A3) after Injection of Humic Materials.

	A1	A2	A3
DOC, ppm	14.5	12.0	50.8
plutonium-239, mBq/L	0.005	0.004	0.041
americium-241, mBq/L	0.015	0.027	0.015
thorium-232, mBq/L	0.37	0.47	0.63
uranium-238, mBq/L	0.75	0.71	1.52
radium-226, mBq/L	1.04	1.59	0.48
radium-228, mBq/L	2.29	3.18	4.66
calcium, ppm	9.6	9.5	12.4
magnesium, ppm	0.93	0.76	0.93
strontium, ppb	53	59	61
aluminum, ppm	<0.10	<0.10	0.25
iron, ppm	0.79	8.38	6.02
silica, ppm	3.10	3.92	2.25

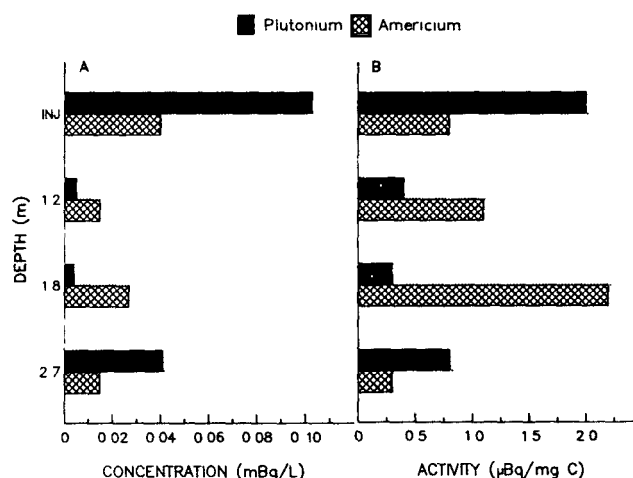


Figure 5. Actinide levels found in the injection and breakthrough waters. (A) Total concentrations of $^{239,240}\text{Pu}$ and ^{241}Am in injection water (INJ) and breakthrough samples at three depths. (B) $^{239,240}\text{Pu}$ and ^{241}Am concentrations normalized to the total amount of DOC in each sample.

injection and were primarily fulvic acids. Therefore, fulvic acids containing the largest concentration of free carboxylic functional groups travel fastest in the aquifer.

Trace metal and radionuclide concentrations at each depth after injection are reported in Table V and shown graphically in Figures 5–7. The synthetic actinides, plutonium and americium, in the absence of an industrial source occur in the environment through fallout from nuclear weapons testing and are associated with organics in the surface water before injection. In the breakthrough waters, the total concentration of both actinides decreased as the DOC is retarded in the subsurface (Figure 5A). However, if these concentrations are normalized to the amount of mobile DOC present (<30 000 MW) in injection and breakthrough waters (Figure 5B), an enhancement is revealed for americium. The smaller, mobile humics have therefore become enriched in americium, the only source being the larger, less mobile colloids.

This is in agreement with laboratory studies which show americium interactions with humic materials overwhelm those with inorganic colloids and silica (29). Furthermore, this enrichment appears to be a function of residence time in the aquifer. The largest enhancement is seen at 1.8 m, where the flow rate of injected water was slowest (178 cm^3/min). In contrast, at 2.7 m, the depth with the fastest flow (636 cm^3/min), no enhancement was observed. The

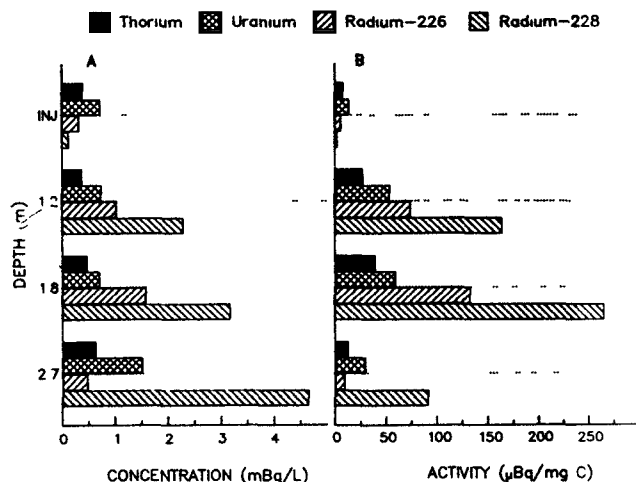


Figure 6. Uranium, thorium, and radium levels found in injection and breakthrough waters (A) Total concentrations of thorium (232), uranium (238), radium (226), and radium (228) in injection water (INJ) and breakthrough samples at three depths. (B) Concentrations of radionuclides normalized to the total amount of DOC in each sample

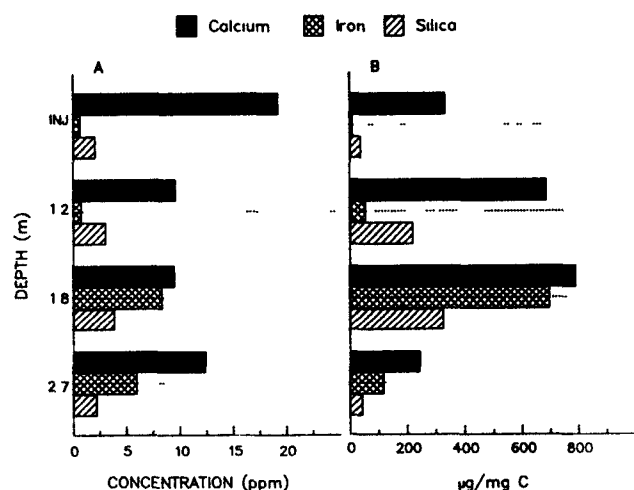


Figure 7. Calcium, iron, and silica levels found in the injection and breakthrough waters (A) Total concentrations of calcium, iron, and silica in injection water (INJ) and breakthrough samples at three depths (B) Concentrations of trace metals normalized to the total amount of DOC in each sample

1.2-m layer, with an intermediate flow rate ($477 \text{ cm}^3/\text{min}$) shows an intermediate enhancement. This observation supports previous studies of dissociation kinetics of rare earth metals to humics which have suggested that flow rate may be an important factor in transport (1). In contrast, plutonium shows no such enhancement with carbon concentrations, because of the intractable nature of $\text{Pu}(\text{OH})_4$ when sorbed to the larger colloidal particles (30).

Total concentrations of thorium, uranium, and radium in these samples are shown in Figure 6A. Total radium concentrations were elevated over those in the original injection water at all sample depths. In addition, thorium and uranium concentrations were higher at the 2.7-m depth. When the concentrations are normalized to DOC values (Figure 6B), a dramatic enhancement over pre-injection values is apparent at all levels. Again, the mobile humics ($<30,000 \text{ MW}$) have become enriched in these radionuclides.

Breakthrough patterns have been studied at this site by using chlorides as a conservative tracer. These results,

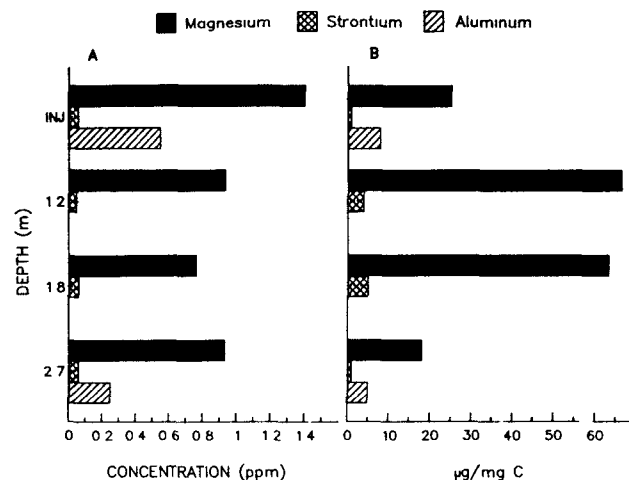


Figure 8. Magnesium, strontium, and aluminum levels found in the injection and breakthrough waters (A) Total concentrations of magnesium, strontium, and aluminum in injection water (INJ) and breakthrough samples at three depths (B) Concentrations of trace metals normalized to the total amount of DOC in each sample

reported elsewhere (18–21), demonstrated little mixing of injected water with the background aquifer waters. The source of this increase in radionuclide concentrations must therefore be the aquifer sands (see Table II). The natural organics, predominantly fulvic acids, which are very high in carboxylate content, apparently mobilize the radionuclides from the aquifer matrix. In keeping with this hypothesis, the enhancement of radionuclide concentrations normalized to mobile carbon (Figure 6B) also shows a correlation to residence time in each layer (i.e., $1.8 \text{ m} > 1.2 \text{ m} > 2.7 \text{ m}$).

The concentrations of selected trace metals in the breakthrough waters are presented in Figures 7 and 8. Iron, silica, calcium, and magnesium show the same patterns as the natural radionuclides and, therefore, are being mobilized in the same manner. However, even though it is a major constituent of the sands, aluminum appears to be resistant to mobilization, possibly because of its tendency toward hydrolysis under these conditions.

Conclusions

Injection of natural organic materials into a shallow, sandy aquifer has demonstrated the ability of low molecular weight, high carboxylic content fulvic acids to mobilize and transport trace metals and radionuclides in subsurface environments. The fallout-derived radionuclides, plutonium and americium, associated with the organic surface waters move with the small organics during forced injection in the subsurface. Americium apparently moved from the less mobile colloidal materials to the smaller, more mobile fulvics during transport. This effect increases with decreasing flow rate in the aquifer. Plutonium, on the other hand, did not reequilibrate onto the smaller materials, consistent with its stronger binding properties and hydrolysis constants.

The natural radionuclides, thorium, uranium, and radium, are found in both the injection water and in the subsurface aquifer sands before the forced injection experiment. Their concentrations in the subsurface waters were enhanced by the injection of small molecular weight organics in the aquifer. This effect also increased with decreasing flow rate. The elevated concentrations of the

natural radionuclides indicate that the natural organics are capable of mineral dissolution and leaching of these radionuclides on reasonably short time scales and under ambient conditions

The results obtained in this study are relevant to the possible transport of radionuclides from nuclear waste repositories by natural humic and fulvic acids. In surface or subsurface conditions of sufficiently low temperatures, where natural organics of low molecular weight and high carboxylate content can be present, such species could increase dissolution from containment matrices and enhance the mobility of radionuclides. Indeed, preliminary studies in this laboratory have indicated that fulvic and humic acids can etch borosilicate glass and can leach uranium from doped glass matrices (31). It will be important to perform further evaluations on the abilities of natural humic and fulvic acids to interact with low-level radioactive waste under planned storage and containment strategies. In addition, it should be noted that these natural chelating agents or synthetic analogs might prove useful in the mobilization of radionuclides under conditions where that would be advantageous in site cleanups.

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